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ADVANCES IN HIGH-POWER CHEMICAL LASERS

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Advances in High-Power Chemical Lasers

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Abstract Chemical laser is characteristically energized by an exoergic chemical reaction which produces the necessary population inversion of lasing species. Based on the enormous chemical energy deposit, several kinds of chemical laser now deliver the cw power outputs of megawatt magnitude, far higher than any other lasers. Accordingly, along with the improvement of gas medium homogeneity and mode-selectivity of the optical cavity, chemical laser has also attained high beam quality of a near diffraction-limited divergence angle. Recent efforts in developing new chemical laser have focused on the short wavelength chemical laser system, e. g. from the early DF ($\lambda = 3.8\,\mu m$) and HF ($\lambda = 2.7\,\mu m$) to late $O_2(^1\Delta)$-I ($\lambda = 1.315\,\mu m$) and HF overtone transition laser ($\sim 1.35\,\mu m$). At present the community of chemical physics are much interested in the visible chemical laser which may be realized by stimulated emission of an electronic excited state.

Key words chemical laser, HF/DF chemical laser, $O_2(^1\Delta)$-I chemical laser

I. A Special Member in the Large Laser Family

Among laser devices, the chemical laser is relatively less well-known. The familiar gas, solid-state, and semiconductor

laser devices, as well as the newly-developed (in recent years) free-electron laser and X-ray laser devices, are basically in the realm of physics. Only the chemical laser device is a product typically combining chemistry and physics. As early as 1961, not long after the appearance of the world's first ruby laser device, a Nobel Prize winner (for chemistry), J. C. Polanyi, proposed the concept of the chemical laser. At that time, he had conducted research on the infrared chemical-light-releasing reaction of halogen atoms and hydrogen molecules, thus discovering that the reactive nascent hydrogen-halide molecules exhibit population inversion at the vibrational-rotational energy levels. This is the fundamental principle of forming chemical lasers. On this basis, Pimental's team was successful in their research on the world's first hydrogen-chloride chemical laser device. Different from any other types of laser devices, the chemical laser device utilizes chemical energy for pumping. Here, the author has in mind only the narrowly defined chemical laser, not including such chemical lasers, in the broad sense of the term, as the kind of photolytic CF_3I device. Because of the enormous energy stored in chemicals, the chemical laser device can possibly achieve very high power output without a enormous power source. Several years ago, at the White Sands Guided Missile Range the United States Air Force built a neon-fluoride chemical laser device called MIRACL; its continuous wave output power is as high as 2.2MW, two orders of magnitude higher in output power than the carbon dioxide laser device excited by electricity, the higher power

output attained to date. As a type of high power laser, in addition to high average power there should have good light beam quality. In this respect, the chemical laser also has its advantage. Since the working medium of a chemical laser device flows at a high velocity, excess heat is continuously being carried away. Thus, there will not be the problem of thermally induced optical distortion as in the case of a high-power solid-state laser device. In another aspect, because of low density and fast diffusional blending in the working medium of the chemical laser device, the homogeneity of medium refractivity is much better than the conventional electrically-excited gas laser device. As reported, given the operating conditions of MIRACL device with its mean power output greater than 1MW, light beam quality approaches a twofold diffraction limit value.

II. Fundamentals of Chemical Laser Device

During the development of chemical laser devices in the past two decades, generally there were the following steps: experiments on the theoretical principle, exploration of new systems, and development of practical high-power devices. Progress at each step was inseparable from research on molecular reaction dynamics. We know that in the working-medium systems in all available chemical laser devices, with the exception of oxygen-iodine chemical laser device, all the excited particles are vibrational-rotational excitation-state molecules. The most

important condition is that the molecules of the chemical reaction products have very high vibrational-rotational population inversion. This not only requires fast reaction speed and high energy release in addition to the fact that the most chemical energy is converted into molecular vibration, not heat energy; moreover, the collision-quenching relaxation rate of the vibrational-excitation state should be as small as possible. Moreover, these states are related to working temperature, pressure, excitation method, as well as species and densities of the quenching impurities. On these problems, research on molecular reaction dynamics can provide much information. Up to the present, scores of types of chemical laser devices were built with excited molecules (as HCl, HBr, HF, DF, CO, F_2, CO_2, I and OH) and output wavelengths from 1.315 to 10micrometers. Since there are many vibrational and rotational energy levels of molecules, when there is an absence of frequency-selected elements in the resonant cavity, all outputs are in multiple spectral lines. Among these chemical laser systems, HF/DF and $O_2(^1\Delta)$-I are the two types that are the most hopeful candidates currently for high-power devices; both types are also the focus of research.

III. Two Most Hopeful Chemical Laser Devices: Combustion-Driven HF/DF Laser and Oxygen-Iodine Energy-Transfer Laser

Since approximately the early seventies, several countries, including the United States and the former Soviet Union, secretly began development of combustion-driven HF/DF laser devices; up to the eighties, these developments were on a relatively large scale. In the United States, as an option for the advanced Strategic Defense Initiative, the MIRACL DF laser device and ALPHA HF laser device were built. The power output of the latter was as high as 5MW, on a continuous wave basis. In the ALPHA HF device, F_2 (or NF_3) reacts with H_2 (or D_2, CS_2, hydrocarbons, among other chemicals) in a combustion chamber to generate high temperatures over 1500K to dissolve F_2 or NF_3 to form F atoms. The combustion products are jetted out from ultrasonic array nozzles for expansion and temperature reduction at a Mach number as high as 4. At the nozzle exit, one adds H_2 (or D_2) to react with F atoms to produce HF· (or DF·) vibrationally-excited molecules. By the action of the optical resonant cavity, excitation emission is generated. The HF laser output is composed of branch P of more than 10 spectral lines from the vibration transition (3-2), (2-1) and (1-0); the wavelengths are in the range 2.5 to 3.0micrometers. Besides the above-mentioned vibrational regions for the DF laser output, there is the (4-3) region with wavelengths in the range 3.7 to 4.0micrometers. The gain of the HF/DF laser is relatively high; by using an nonsteady-state cavity to select oscillations in the fundamental mode, light-beam quality approaches the diffraction limit. Technically, the HF/DF laser devices have matured, but with some

shortcomings, such as higher wavelength and higher toxicity in fluorine. Table 1 lists some operating conditions of HF, DF and oxygen-iodine chemical laser devices (with combustion driving) and laser output performance.

Invention of the chemical oxygen-iodine laser (COIL) occurred about a decade later than HF/DF laser. The principle of COIL differs most decidedly from other chemical lasers. First, hydrogen peroxide (from alkali) solution reacts with chlorine to generate substeady unit-weight $O_2(^1\Delta)$; the latter collides with iodine atoms for energy transfer to excite iodine from its ground state I (^2P 3/2) to I ($^2P_{1/2}$) spinning-rail excited state in order to make an iodine atomic laser operating at 1.315micrometers. The chemical dynamic process of the $O_2(^1\Delta)$ iodine energy transfer laser device can be expressed with the following equations:

(1) Occurrence of singlet-state oxygen

$$Cl_2 + 2H_2O_2 + 2MOH \rightarrow O_2(^1\Delta) + 2H_2O + 2MCl$$

In the equation, M=K,Na,Li

(2) Generation of iodine atoms

$$I_2 + nO_2(^1\Delta) \rightarrow 2I + nO_2(^3\Sigma), \quad 3 \leqslant n \leqslant 5$$

(3) Singlet-state oxygen-iodine energy transfer

$$O_2(^1\Delta) + I(^2P_{3/2}) \rightarrow I^*(^2P_{1/2}) + O_2(^2\Sigma) + \Delta E, \quad \Delta E = 279\ cm^{-1}$$

(4) Energy accumulation reaction of the excited-state oxygen and iodine

$$O_2(^1\Delta)I^*(^2P_{1/2}) \rightarrow O_2(b^1\sum) + I(^2P_{3/2})$$

(5) Quenching reaction of excited-state iodine

$$I^*(^2P_{1/2}) + M \rightarrow I(^2P_{3/2}) + M, \quad M = H_2O, I, H_2O_2, Cl_2 \quad \text{etc.}$$

At present, subsonic COIL devices with a continuous-wave 1kW output were built in the United States, Japan, Russian and China; however, a US Air Forces weapons laboratory has constructed an ultrasonic device with a continuous-wave output of 35kW, called ROTOCOIL; its light-beam quality has achieved the level of "very good." The key sector of this technology is the structure of the optical cavity. Since COIL has a small signal gain, in only in the thousandths per centimeter, with a large volume of excitation medium, thus a less-than-desirable effect can be obtained by using a steady-state cavity or a conventional nonsteady-state cavity. This shortcoming may be overcome by using some new type of optical cavity such as the UR-90 (ring-shaped with 90^0 rotation) or the VRM cavity (variable-reflectivity nonsteady-state cavity). Another important key technique of the COIL is its highly efficient singlet-state oxygen generator, which is the energy source of the COIL. Although the lifetime of $O_2(^2\Delta)$ is as long as 45min, however, it quenches rapidly in solution. To reduce such losses, the contact time between $O_2(^1\Delta)$ and sclution should not be long. However, if the contact time is too short, the reaction with chlorine is incomplete. To solve this dilemma,

the gas-liquid interface should be as large as possible in the generator with the smallest volume. Experiments are underway on generators of various structures, such as bubble-blowing type, rotating-plate type, and atomizing type. Possibly, the effect of the rotating-plate type is better. Generally speaking, although the development of COIL is not as mature as that of the HF/DF laser device, yet there are advantages of shorter wavelength and lower toxicity. These advantages are very beneficial in its industrial promotion and applications. A 1.315micrometer laser can be transmitted via an optical-fiber cable. At the Kawasaki Heavy Industries Corporation in Japan, a 1kw demonstration device was constructed, transmitting its laser beam through a optical-fiber cable to many processing workshops.

IV. Future of Chemical Laser

With respect to researchers on the chemical laser internationally, on the one hand they are vigorously upgrading available laser devices to achieve practicality; on the other hand, they are actively exploring new chemical laser systems, in particular, visible-light chemical laser systems. Efforts at developing visible-light chemical lasers have not really succeeded. The first difficulty is that the chemical energy release relating to the pumping reaction should be quite high. For example, for green light at 200nm, the reaction energy release should be greater than 240kJ per gram mole. However, the

Table 1 Typical operation parameters and performances of cw supersonic HF, DF and O_2 ($^1\Delta$)-I chemical laser

	HF	DF	$O_2(^1\Delta)$-I
Conditions of the plenum			
Pressure (atm.)	1～5	1～5	0.05～0.1
Temperature (K)	1300～2000	1300～2000	Room temp.
Nozzle area ratio (A/A^*)	13～18	13～18	2
Cavity gas pressure (Pa)	530～1060	530～1060	270～530
Mach number of the supersonic flow (M)	4	4	2
Laser wavelength (μm)	2.7	3.8	1.315
Small signal gain coefficient (cm^{-1})	0.1	0.02	0.008
Optical cavity	Confocal unstable resonator	Confocal unstable resonator	Ring unstable resonator
Maximum power output (W)	1×10^6	2.2×10^6	3.5×10^4
Specific power (J/G)	160～200	100～150	200～240
Chemical efficiency	12～14	12～14	14～37
Output beam quality	Near diffraction-limited	Near diffraction-limited	Excellent

more difficult issue is that the reactant should have its population in the electronic excitation state. Generally speaking, only the amount of energy in the electronic excitation state is sufficient to radiate visible light. For most chemical reactions, however, the released energy is converted into

vibrational, rotational and translational energy of molecules with very little probability of generating electronic excitation state owing to limitation by the distribution function. Nonetheless, light at the end of the tunnel is visible after many years' difficult but persistent explorations by chemists. For example, in the reaction: Na_3+Cl, Na_2^*, the electronic excitation state (with emission of green light at 527nm) has been obtained. As reported, the gain coefficient is 1percent per centimeter. IF, NF, BiF and PbN_3 are among the systems being explored. Recently, an interesting discovery was reported: red fluorescence radiation was observed in $O_2(^1\Delta)$ gas current. The radiation is the radiation of the O_4^* quasi-molecule, as suggested by Japanese scientists. Researchers are hoping to build a visible-light chemical laser by relying on the above-mentioned radiation. What particles can release light?-- this subject is being researched competitively by scientists in Japan, the United States, China, France and Russia. At present, the radiation has been basically determined as radiation from copper chloride; this research fervor has cooled down.

In the international competition in this high-technology realm (chemical laser), China's chemists have also actively participated with some successes. In 1966, the year after Pimental first succeeded with the chemical laser, a team at the Dalian Institute of Chemical Physics (of the Chinese Academy of Sciences) succeeded in observing chemical laser pulses from

hydrogen chloride in a laboratory. In the years that followed, there were the following achievements: a CO chemical laser triggered by electrical discharges, combustion-driven continuous-wave HF/DF laser, HF pulsed laser triggered by electron beam or light, as well as pulsed and continuous oxygen-iodine laser, among others. In addition, research activities relating to molecular reaction dynamics have already recorded advances. In this leading-edge region of technology, China's scientists have some research achievements (such as electrically-triggered pulsed oxygen-iodine laser, and high-power continuous-wave oxygen-iodine laser), entering world-class rankings.

Refer to page 443 of this issue; the author is a member of the editorial committee of this publication, Zhongguo Jiguang [Lasers in Chemistry]. The paper was received for publication on 8 November 1993.

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